

**Supplementary Information: charge regulation of nonpolar colloids**

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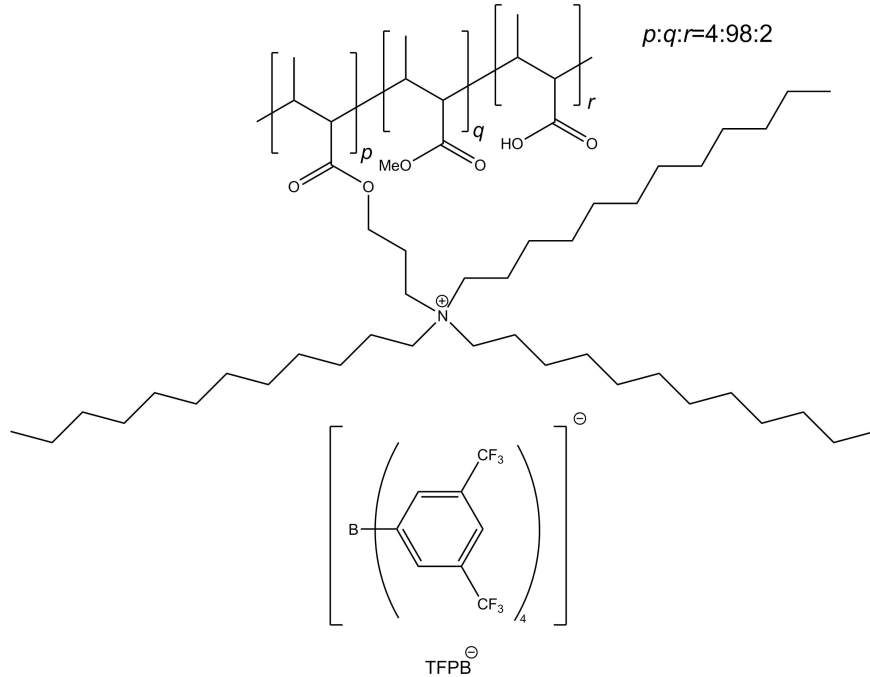
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## I. IONIC-MONOMER MODIFIED NP3 NANOPARTICLES

Nanoparticles NP3 were synthesised by a dispersion copolymerization of methyl methacrylate, methacrylic acid, and the ionic monomer *n*-tridodecyl-propyl-3-methacryloyloxy ammonium tetrakis [3,5-bis (trifluoromethyl) phenyl] borate ([ILM-(C<sub>12</sub>)<sup>+</sup>][TFPB]<sup>-</sup>) in weight ratios *p:q:r* of 4:98:2. The molecular structure of the resulting copolymer is illustrated in Figure 1 where the positive quaternary ammonium ions are covalently bound within the core of the nanoparticle, and the [TFPB]<sup>-</sup> ions are free to dissociate into the bulk.

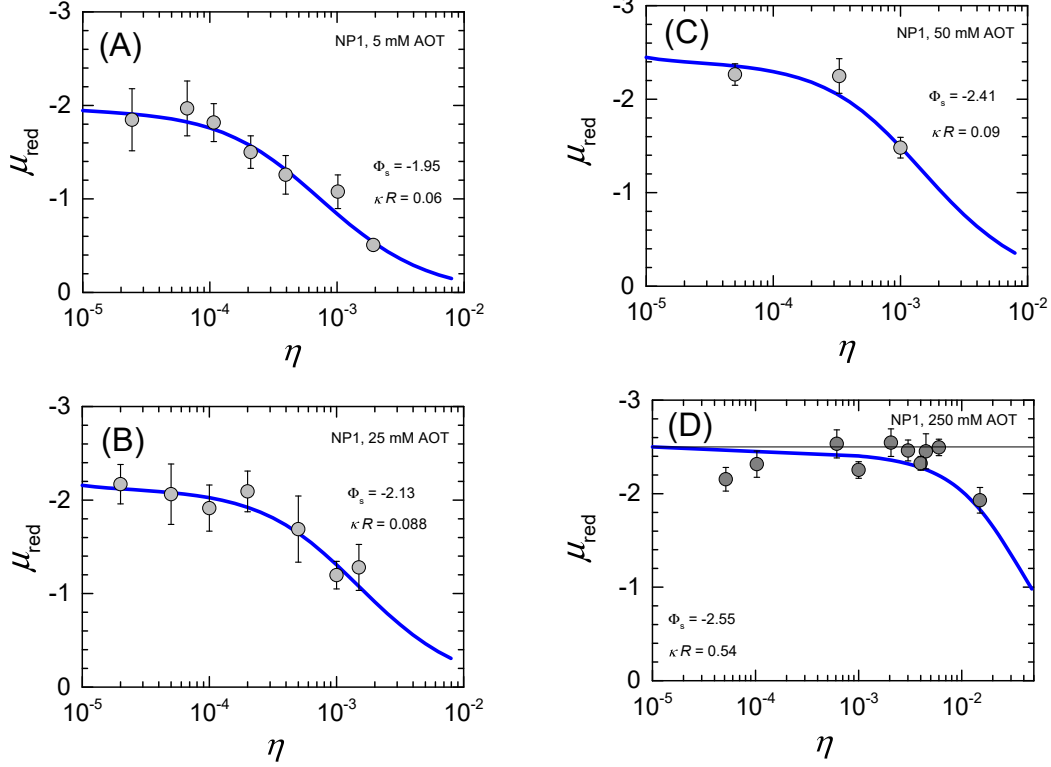


**Figure 1:** Schematic molecular structure of polymeric NP3 nanoparticles.

## II. VARIATION OF MOBILITY WITH ADDED ELECTROLYTE

The effect of the concentration of the surfactant AOT on the packing-fraction dependent electrophoretic mobility  $\mu_{\text{red}}(\eta)$ , measured in dispersions of negatively-charged NP1 nanoparticles, is illustrated in Figure 2. Adding aerosol-OT reverse micelles has two effects: (a) we increase the (negative) scaled particle potential  $\Phi_s$  and, (b) by adding more ions to the dispersion we screen the electrostatic interactions between particles and reduce the dependence of the electrophoretic mobility on the nanoparticle packing fraction  $\eta$ . These trends are apparent from a quick comparison between the mobilities (represented by the

circles) measured for  $C_{\text{AOT}} = 5 \text{ mmol dm}^{-3}$  (shown in Figure 2(a)) and the data recorded at  $C_{\text{AOT}} = 250 \text{ mmol dm}^{-3}$  (shown in Figure 2(d)). The Kuwabara cell model and a constant potential (CP) boundary condition reproduces these trends very well, as shown by the solid lines in Figure 2.



**Figure 2:** The reduced electrophoretic mobility  $\mu_{\text{red}}$  as a function of volume fraction in dispersions of NP1 nanoparticles at (a)  $C_{\text{AOT}} = 5 \text{ mmol dm}^{-3}$ ; (b)  $C_{\text{AOT}} = 25 \text{ mmol dm}^{-3}$ ; (c)  $C_{\text{AOT}} = 50 \text{ mmol dm}^{-3}$ ; and (d)  $C_{\text{AOT}} = 250 \text{ mmol dm}^{-3}$ . The solid lines are mobilities calculated using a Kuwabara cell model assuming a constant potential (CP) boundary condition at the surface of the particle. The fitted surface potentials  $\Phi_s$  and screening parameters  $\kappa R$  are reproduced in Table II of the paper.